Science of the Total Environment 630 (2018) 931–942

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

The long-term impacts of anthropogenic and natural processes on groundwater deterioration in a multilayered aquifer

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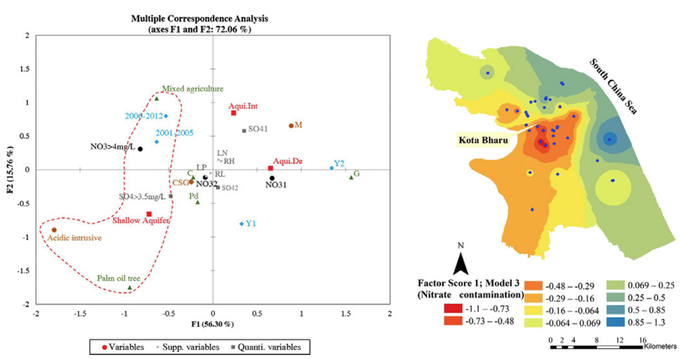
HIGHLIGHTS

- Controlling factors in groundwater chemistry vary in the multi-layered aquifer.
- In shallow aquifer, Nitrate is controlled by Palm trees and agricultural cultivations.
- In the intermediate aquifer, salinity is associated with marine deposits.
- Nitrate shows increasing trend in shallow in Kelantan from 1989 to 2012.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 20 September 2017
Received in revised form 15 February 2018
Accepted 16 February 2018
Available online xxxx

Editor: R Ludwig

Keywords:
Groundwater chemistry
Natural processes
Anthropogenic factors
Land use
Multiple correspondence analysis

ABSTRACT

In many regions around the world, there are issues associated with groundwater resources due to human and natural factors. However, the relation between these factors is difficult to determine due to the large number of parameters and complex processes required. In order to understand the relation between land use allocations, the intrinsic factors of the aquifer, climate change data and groundwater chemistry in the multilayered aquifer system in Malaysia's Northern Kelantan Basin, twenty-two years hydrogeochemical data set was used in this research. The groundwater salinisation in the intermediate aquifer, which mainly extends along the coastal line, was revealed through the hydrogeochemical investigation. Even so, there had been no significant trend detected on groundwater salinity from 1989 to 2011. In contrast to salinity, as seen from the nitrate contaminations there had been significantly increasing trends in the shallow aquifer, particularly in the central part of the study area. Additionally, a strong association between high nitrate values and the areas covered with palm oil cultivations and mixed agricultural have been detected by a multiple correspondence analysis (MCA), which implies that the increasing nitrate concentrations are associated with nitrate loading from the application of N-fertilisers. From the process of groundwater salinisation in the intermediate aquifer, could be seen that it has a strong correlation the aquifer lithology, specifically marine sediments which are influenced by the ancient seawater trapped within the sediments.

1. Introduction

The spatial and temporal characteristics of groundwater often reflect the natural and anthropogenic activities which surround it. During the

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https://doi.org/10.1016/j.scitotenv.2018.02.190

last few decades, the natural ecosystems, including forest, grassland, aquatic, and desert ecosystems, have been altered more extensively and rapidly, compared to other points in time due to large and massive land use and land cover changes (Muñoz-Rojas et al., 2015). By year 2000, around 55% of the earth's biomes had been converted into pastures, agricultural lands, settlements and other land uses (Salazar et al., 2007). Furthermore, it is predicted that the area land under crop cultivation in developing countries may have increased to 110 million ha by 2050 (FAO, 2006). The pattern of land use in drainage basins is one of the most important human-induced driving forces, due to the rapid population growth, development of agriculture, industry, and urbanisation which impact the hydrogeological cycle in terms of quality and quantity (Hao et al., 2008; Levashova et al., 2004). Therefore, the types, intensity, and frequency of land use activities can impact on water bodies and degrade water quality (Meneses et al., 2015). The declining groundwater quality increase the concerns about the effects posed on groundwater ecosystems by the anthropogenic processes (Hancock et al., 2005).

The chemical composition of groundwater could be influenced by several processes, such as weathering, dissolution, precipitation, absorption, ion exchange, and the oxidation/reduction processes (Jeong, 2001). However, the influences posed by natural processes and anthropogenic input are not easy to distinguish solely based on the groundwater chemical composition and constituents. Basic geochemical information on the groundwater and the controlling mechanism of water quality could be obtained from the major cations and anions in the groundwater (Andrade and Stigter, 2011). However, detecting and understanding the past and current trends in groundwater chemistry are the key factors in discovering the main mechanism. The mechanism occurs where the locations of the increasing pollution are regulated. Knowledge of the mechanism and its location greatly assists management authorities and policy makers in their efforts to take appropriate restoration measures (Abaurrea et al., 2011).

Determining the factors controlling groundwater quality is very challenging in complex hydrogeological systems, especially in multilayered aquifers. Moreover, it is possible that the controlling factors themselves vary over time, due to the changes taking place in the hydrological and climatological conditions, and anthropogenic activities. In the recent decades, several studies have considered the effects of land use changes on water systems (Narany et al., 2017; Heißerer et al., 2016; Ovalle et al., 2013; Andrade and Stigter, 2011). However, the number of studies focusing on the long-term changes of land use, particularly those associated with complex aquifer systems, is very limited. With concerns regarding the impacts of the rapid increase of human activities on groundwater quality, the following questions have arisen: (i) Have any discernible groundwater hydrochemistry changes occurred during the investigation? (ii) Which factors (natural or anthropogenic processes) are the primary drivers of changes in groundwater chemistry in this complex aquifer system? (iii) Can the dominant factors controlling groundwater quality be changed? (iv) Are there any distinguishable relationships between human activities and groundwater quality? (v) Which land use categories poses significant effects on groundwater quality?

An effort was done by the present study in finding the answers to the above-mentioned questions. This was with the purpose of evaluating the impacts of long-term human activities, and the climatological and hydrogeological conditions on the factors controlling groundwater chemistry in the multilayered aquifer located in Malaysia's in the Northern Kelantan Basin. This location is where rapid deforestation and the extension of agricultural lands (around 7%) had been reported from the 1970s until the 1990s (Hassan, 2004). In order to achieve this objective, the application of the multi method approach was performed by integrating of the geochemical methods and the time series analysis. With this, the factors controlling the groundwater hydrochemistry could be determined and the evaluation of groundwater quality trends from 1989 to 2012 in the multilayered aquifer system in Northern Kelantan could be evaluated. In addition, more advanced methods, such as multiple correspondence analysis (MCA) and geospatial modeling, were implemented in order to construct explanatory models between quantitative and qualitative parameters. Apart from that, groundwater hydrochemistry, lithology, land use activities, climatology, groundwater level, and years of monitoring in the study area fall under these parameters. The results obtained from this study can serve as beneficial references in improving the groundwater resource management in rapidly developed areas, where massive human activities take place.

2. Materials and methods

2.1. Study area

The Northern Kelantan Basin has covered an area of approximately 880 km^2 on the north-east coast of Peninsular Malaysia (Fig. 1). This area is characterized by the low elevation coastal plain which stretches at approximately 60 km in length and 24 km in width along the coast. It faces the South China Sea where a fertile alluvial plain is located, making it ideal for agricultural activities. The Northern Kelantan Basin is mainly drained by four major rivers, namely the Kelantan, Pengkalan Datu, Kemasin and Semerak. All of the river flow systems are in accordance to the northwest direction before the river is discharged into the South China Sea.

This area is the most populated area in Kelantan (Said et al., 2013). To illustrate this, the population of this state was around 1,174,000 in 1990. However, from that year it increased to 1,800,000 in 2016 (Kumari and Kavanagh, 1990), which mostly occupied the districts of Kota Bharu, Tumpat, and Bachok in particular. In fact, Kelantan's economic activities are primarily centered on agricultural activities, primarily the cultivation of paddy rice, oil palm, rubber, and tobacco. Despite the significant improvement of the economic status in the state during the last decades, the environmental stability of it has been affected by the intensive agricultural activities and the population growth in the study area. From the 1970s to the 1990s, e Kelantan had been experiencing rapid deforestation (around 7%), and the development of urban and farm areas (Jusoff and Setiawan, 2003). Subsequently, deforestation and unplanned rapid urbanisation have become the primary flood causes in this area. Moreover, intensive agricultural and residential activities put pressure on water quality and quantity as the major source of water supply in the Northern Kelantan Basin (Samsudin et al., 2008).

2.2. Regional climate

Since Peninsular Malaysia is in the equatorial zone, it experiences a tropical rainforest climate, which is controlled by the north-east and south-east monsoon regimes. The mean annual rainfall from 1979 to 2011 was 2563 mm. Throughout this period more than half of the rainfall took place from October to December. It was due to storms occurring in the raining season (Saghravani et al., 2015). According to the Malaysian Meteorological Department's data, there had been a slight increase in the mean annual rainfall from 1989 to 2012 (Fig. 2).

The maximum and minimum amount of rainfall which was amounted to 1689 mm and 3734 mm occurred in 1989 and 1999, respectively. However, the average amount of rainfall which had taken place from 2007 to 2012 was approximately 3210.8, which represented a considerable amount of increase in rainfall during this period. Furthermore, the average temperature had been around 26 °C throughout the year in the area, where the study was conducted. According to Shaffril et al. (2011) Kelantan is one of the four states in the East Region of Peninsular Malaysia which are affected by climate change. This can be seen from how the atmosphere's temperature has increased from 1.75 °C to 2.69 °C in the last 40 years in this region (Tangang et al., 2007), which has resulted in higher reliability in terms of rainfall and severe floods.

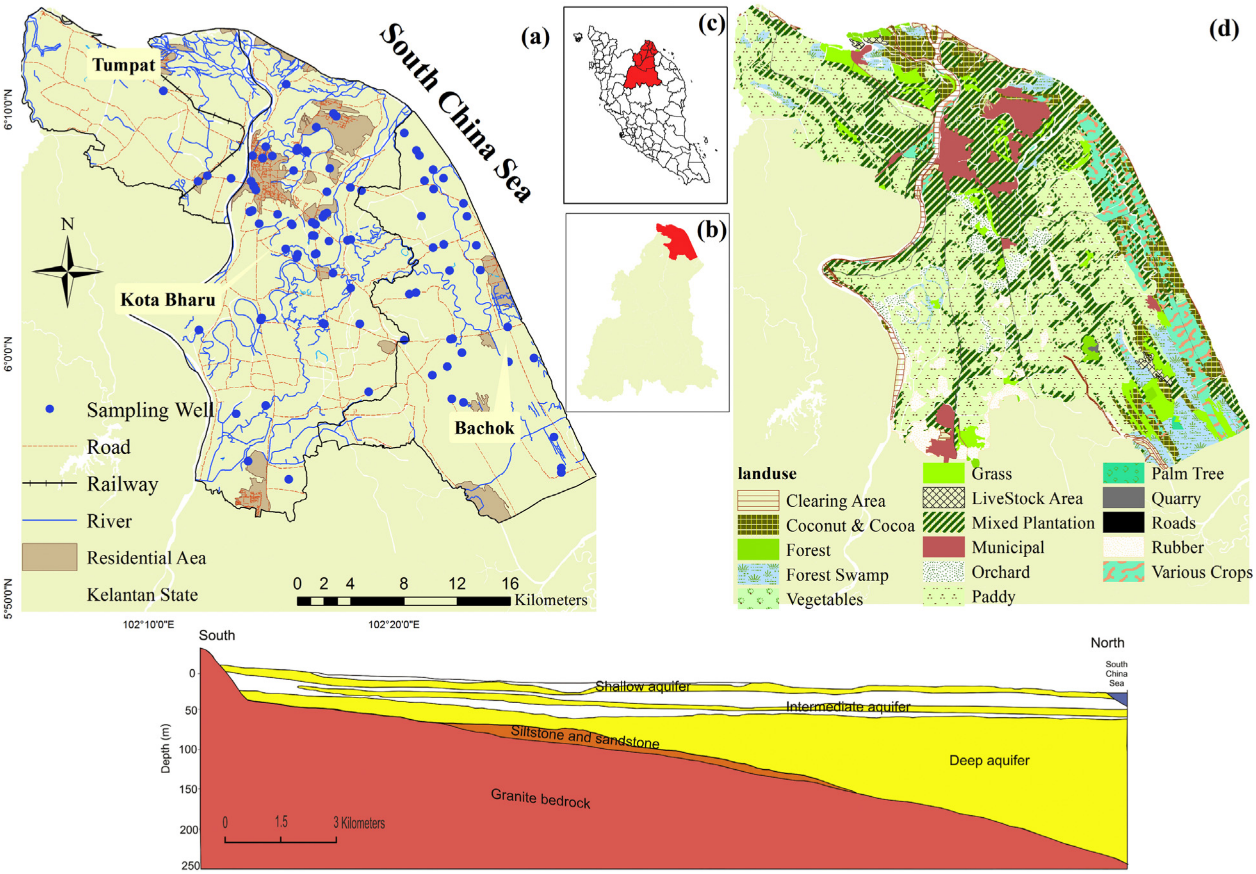

Fig. 1. Location of the Northern Kelantan (a), in Kelantan State (b), Peninsular Malaysia (c), land use activities 2011 (d), and geological cross section (e).

2.3. Hydrogeological sitting

Approximately, 41% of the water production in Kelantan originates from groundwater. The utilization of groundwater in Kota Bharu, Kelantan began in 1935, and from then the usage demand has increased by 2.5% annually; the demand for the production of groundwater in the entire state was estimated to be 164.7 m^3/d in 2010 (Suratman, 2010). In the stated year, Kota Bharu, Bachok, and Tumpat displayed the highest demand for groundwater, which amounted to around 96.82, 28.42, and 22.51 m^3/d respectively (Table 1). However, the groundwater was drawn from the deeper aquifers in some cities, such as Pintu Geng, Tanjung Mas, and Kg. Puteh.

The aquifer system in the Northern Kelantan Basin is formed by the quaternary alluvium deposits, which consist of containing three layers of the aquifer. Besides, the aquifer is characterized by the thickness of the layer, which ranges from 25 m in the inner parts to 200 m at the coastal area (Suratman, 1997).

The shallow aquifer extends to a depth of <20 m, and it extends throughout the entire basin. It consists of light-brown sand with gray clay and shells. The intermediate aquifer lies at a depth of 20–0 m. It is

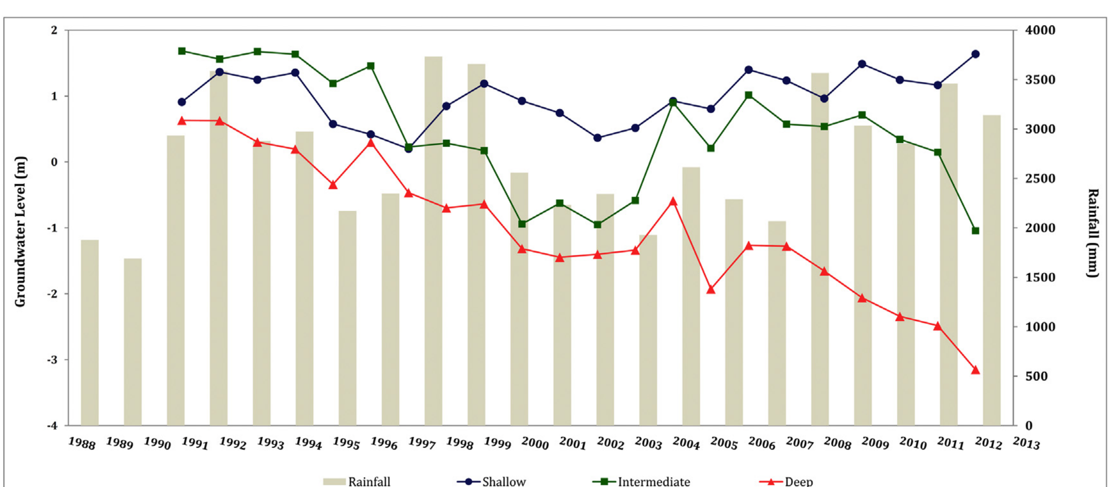

Fig. 2. Average annual rainfall from 1989 to 2012 and average groundwater level in three different aquifer layers in Northern Kelantan Basin.

Table 1
Groundwater use in Kelantan (2010).

District	Population 2010	Groundwater use (%)	Groundwater demand m^3/d
Kota Bharu	509,600	95%	96,824
Bachok	142,100	100%	28,420
Tumpat	173,200	65%	22,516

made up of light gray, medium- to coarse-grained sand. The deep aquifer extends to >50 m. It consists of coarse-grained sand, along with fine- to medium-size gravel. The intermediate and deep aquifers are separated by a layer of impervious, blue-gray clay and shells (Pfeiffer and Tieddemann, 1986). In general, the infiltration into the shallow aquifer is from precipitation runoff, leakage from rivers, and consumer pipe infiltrate. Wastewater consists of nitrogen, organic matter, inorganic salts, heavy metals, and bacteria (Gasana, 2014). Based on the studies conducted by Noor (1979), the shallow and intermediate aquifers are hydraulically interconnected due to the presence of a silty layer between them. Moreover, it was revealed Haryono et al. (1995) and Samsudin et al. (2008) in North Kelantan that the groundwater in the intermediate aquifer extends up to an average distance of 6 km from the coastline. It contains fossil seawater, which was probably trapped during its original sedimentation in this area.

The geological studies in the Northern Kelantan Basin (Macdonald, 1967) have revealed that, although the alluvial plain mainly consists of the quaternary unconsolidated alluvium and marine sediments, there are contents of metamorphic rocks in the bedrocks. According to Saghravani et al. (2015) quaternary sediments in the plain contain silty and clayey lenses, which are inter-bedded within sand and gravel. The upper layer near the coast consists of medium to coarse-grained sand including silty and/or peat lenses.

2.4. Groundwater data

In this study, >1000 groundwater samples were collected from 83 different sampling wells by the Minerals and Geoscience Department Malaysia (JMG). These wells are completed into the shallow, intermediate and deep aquifers. They were installed between 1989 and 2011. Although the monitoring frequency was conducted on a monthly basis, however, the occurrence of unexpected events prevented the sampling of some wells. The groundwater samples were collected from domestic and agricultural wells, with different depths of the sampling screens, which ranged from 3 to 127.5 m.

2.5. Statistical analyses

2.5.1. Trend analysis

The methods utilized for testing the detection of significant trends in hydro-meteorological time series can be classified into parametric and non-parametric methods (Jain and Kumar, 2012). The Mann-Kendall test (Kendall, 1975; Mann, 1945) is a non-parametric test for trend detection in the time series data. Furthermore, this rank-based procedure test is resilient to extreme conditions and good to be used with skewed variables (Partal and Kahya, 2006). The present study applied this test for detecting the significance of trends in annual hydrogeochemical time series data. Based on the test, it was assumed from the null hypothesis (H_0) that no trend or serial correlation over time was shown from the observations. However, it was assumed from an alternative hypothesis (H_1) that there was a significant trend in the time series observations. According to the study, when P value was less than or equal to the significance level ($\alpha = 0.05$), the null hypothesis would be rejected. However, Sen's slope estimator (Sen, 1968) should be applied in order to detect an increasing trend (positive Sen's slope) or a decreasing trend (negative value of Sen's slope) shown in time series observation.

2.5.2. Correspondence analysis and multiple correspondence analysis

The correspondence analysis (CA) is a multivariate descriptive data analytic technique, which is applied for handling and representing the relationship between categorical data (Ayele et al., 2014). The handling and representation are conducted by calculating a similarity matrix of the data which then undergoes a statistical transformation involving its diagonalisation and the extraction of the eigenvectors and eigenvalues (Andrade and Stigter, 2009). This technique provides information, such as the information regarding the diagonalisation of the similarity matrix for the extraction of the factors, which is similar to the principal component analysis (Andrade and Stigter, 2013). In general, the correspondence analysis capable of simplifying complex data and providing a detailed description from both the qualitative and quantitative variables. These actions are done by separating the data into classes, which is an important feature of the multiple correspondence analysis (MCA). Moreover, this technique is the multivariate version of CA, used to analyse the pattern of relationships between several nominal variables (Abdi and Valentin, 2007). Each nominal variable consists of several levels, and each of these levels is coded as a binary variable. For example, a level will be labeled as 1 if samples fall within a variable class, otherwise it will be labeled as 0). Additionally, the principle of the MCA is to extract the first factors which retain the maximum amount of information contained within the similarity matrix. This is followed by the application of the information to the statistical transformation where its diagonalisation and the extraction of the eigenvectors and eigenvalues are involved. These two matters define the explained fraction of the initial data variance (Andrade and Stigter, 2013). One of the advantages of the MCA is its ability of converting a matrix of data into a factor plane, which depicts the rows and columns of the matrix as points. Furthermore, it enables a geometrical representation of all the information (Greenacre and Hastie, 1987). In the current study, the row point corresponded to the observations from sampling wells, while the column points corresponded to the quantitative and qualitative variables. The quantitative variables, including the electrical conductivity namely Na, Cl, SO_4, NO_3, Fe, Ca, and K data, were transformed into ordinal variables by subdividing each of them into three classes. This classification was based on the criterion of equal sample distribution during the preprocessing procedure (Table 2). Moreover, qualitative variables, including the type of aquifer, rainfall data, groundwater level variations, land use, geology and year of sampling were classified based on the details of their definitions (Table 2). In the MCA, the computation of factor planes and their axes were conducted in the order of the most to the least explicative (Greenacre and Hastie, 1987). The current study presents the most explicative factor plane based on the inertia value, which is defined as the total Pearson Chi-square for two-way frequency table, divided by all of the totaled-up observations in the table.

3. Results and discussions

3.1. Spatial and temporal assessment of hydrochemical facies

Hydrochemistry variation can play an important role in understanding the controlling factors that influence groundwater chemistry. The long-term trends in the groundwater quality of the Northern Kelantan Basin have been documented for decades; from the late 1980s to the early 2010s. Furthermore, the patterns of the long-term changes can distinguish between natural and human induced processes which affect groundwater quality in the study area. Piper and Schoeller diagrams were used for the characterization of groundwater chemical compounds between the years 1989 and 2011 in the study area displayed in Fig. 3.

Based on the results of the Piper diagram, Na-HCO_3 and Ca-HCO_3 are the primary chemical compounds of groundwater in both years, which indicates that there had been no significant change observed in the groundwater facies in the study area for over 22 years. However, there was a different case for two groundwater samples in 1989, which

Table 2
Definition of quantitative and qualitative parameters for multiple correspondence analysis (MCA).

Variable	Class	Description	Freq.%
Aquifer	Aqui.Sh	Shallow	37.2
	Aqui.Int	Intermediate	31.7
	Aqui.De	Deep	31.1
Ground water level	LP	Positive	72.3
	LN	Negative	27.7
Rainfall (mm)	RL	<2500	55.8
	RH	>2500	44.2
Year	Y_1	1989–1994	35.3
	Y_2	1995–2000	12.6
	Y_3	2001–2005	27.5
	Y_4	2006–2012	24.5
Land use	G	Forest and grass land	19.2
	C	City	26.3
	MA	Mixed agriculture	26.6
	Pd	Paddy	19.1
	PO	Palm Oil tree	8.9
Geology	M	Marine deposit	22.9
	CSG	Clay, silt, gravel	70.4
	A	Acidic intrusive	6.6
Electrical conductivity (μS/cm)	$COND_1$	≤100	27.1
	$COND_2$	100–185	31.2
	$COND_3$	≥185	41.7
Na (mg/l)	Na_1	≤9	32.6
	Na_2	9–23	32.9
	Na_3	≥23	34.5
Cl (mg/l)	Cl_1	≤8	34.6
	Cl_2	8–34	33.5
	Cl_3	≥34	31.8
SO_4 (mg/l)	SO_4^1	≤1.5	48.7
	SO_4^2	1.5–3.5	26.2
	SO_4^3	≥3.5	25.1
NO_3 (mg/l)	NO_3^1	≤1.5	44.7
	NO_3^2	1.5–4	28.0
	NO_3^3	≥4	27.3
Fe (mg/l)	Fe_1	≤5	35.0
	Fe_2	5–13	35.0
	Fe_3	≥13	30.1
Ca (mg/l)	Ca_1	≤5	35.0
	Ca_2	5–17	34.9
	Ca_3	≥17	30.2
K (mg/l)	K_1	≤4	34.4
	K_2	4–8	33.8
	K_3	≥8	31.8

indicated that groundwater of the Na-Cl type could be represented as a saline water intrusion to fresh water. The groundwater samples which originated from shallow and deep aquifers had a similar anionic composition which was dominated by the HCO_3 ion, with in an abundance order of HCO_3 > Cl > SO_4 (meq/L) and the cationic composition was dominated by the Na ion, with abundance orders of Na > Ca ≥ Mg (meq/L) in 1989. In contrast to shallow and deep aquifers, the intermediate aquifers showed a different ionic composition that was dominated by the Cl ion, with an abundance order of Cl > HCO_3 > SO_4 (meq/L), while the cationic composition was dominated by Na > Ca > Mg (meq/L) in 1989. Although the patterns of the anionic composition of groundwater samples in shallow and deep aquifers in 2011 were similar to the patterns of the samples in 1989, their cationic composition was different, which was dominated by the Ca ion with abundance orders of Ca > Na > Mg and Ca > Mg > Na in shallow and deep aquifers, respectively. Furthermore, the groundwater samples in the intermediate aquifer in 2011 had a similar cationic composition with the samples in the intermediate aquifer in 1989, which were dominated by an abundance order of Na > Mg > Ca. However, their anionic composition was dominated by the HCO_3 ion, with an abundance order of HCO_3 > Cl > SO_4. Therefore, most of the groundwater samples showed Na-HCO_3 and several Na-Cl types in 1989, which slightly changed to Ca-HCO_3 and Na-HCO_3 in 2011. Although, there was no significant difference detected in the groundwater of these types between 1989 and 2011, the differences between the groundwater types in the aquifer layers were indicated in the Piper and Schoeller diagrams. Moreover, the development of groundwater quality index (GQI) specific to freshwater-seawater mixing index (GQI $_{piper.mix}$) and another index (GQI $_{piper.dom}$) can be spatially analyzed to the visual distribution of water quality index through Geographic Information System (GIS) (Tomaszkiewicz et al., 2014). Therefore, Piper domain indexes could be used for a detailed investigation on the groundwater types in different aquifers using the GQI $_{(piper.mix)}$ and GQI $_{(piper.dom)}$ indices in the study area (Fig. 4a). In addition, the diamond filed of the Piper diagram can be divided into six differing domains: I, II, III, IV, V, VI representing $CaHCO_3$, NaCl, mixed $CaNaHCO_3$, mixed CaMgCl, $CaCl$, and $NaHCO_3$, respectively. It was shown from the results that the GQI $_{(piper.mix)}$ varied from a maximum 85.4 to a minimum 15.9 in the shallow aquifer. As for the intermediate aquifer, it varied from a maximum 80.5 to a minimum 12.8 and from a maximum 75.5 to a minimum 30.6 in the deep aquifer. It was also indicated from the results that the evidence of seawater intrusion to groundwater in some samples belonging to the shallow and intermediate aquifers was proven.

The GQI (piper.dom) changed from a maximum 80.9, 84 to a minimum 29 in the shallow aquifer, a maximum 84 to a minimum 27.8 in the intermediate aquifer, and a maximum 82.2 to a minimum 29 in the deep aquifer, respectively. Based on the Piper domains results (Fig. 4a), the majority of the groundwater samples in the shallow aquifer (85.7%) consisted of groundwater types I and III (indicating that each

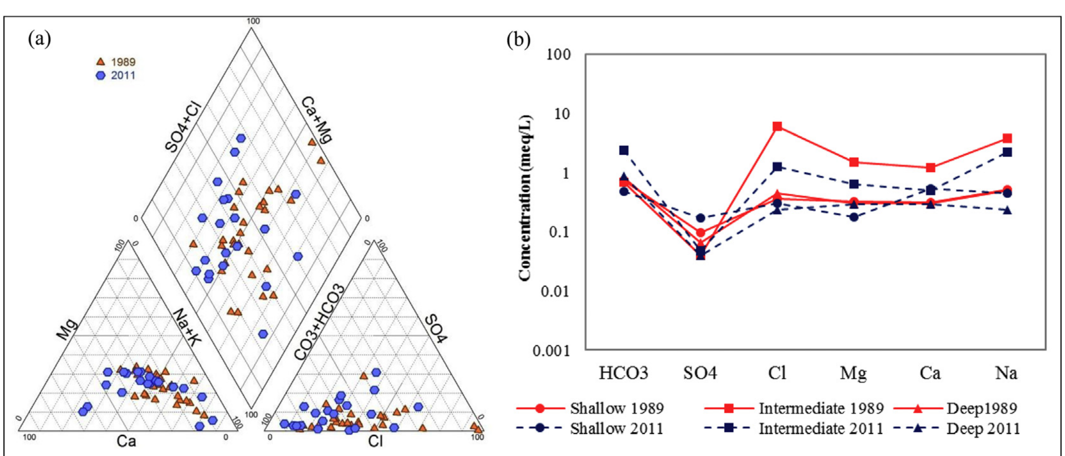

Fig. 3. Piper (a) and Schoeller diagrams (b) of groundwater samples in shallow, intermediate and deep aquifers in 1989 and 2011.

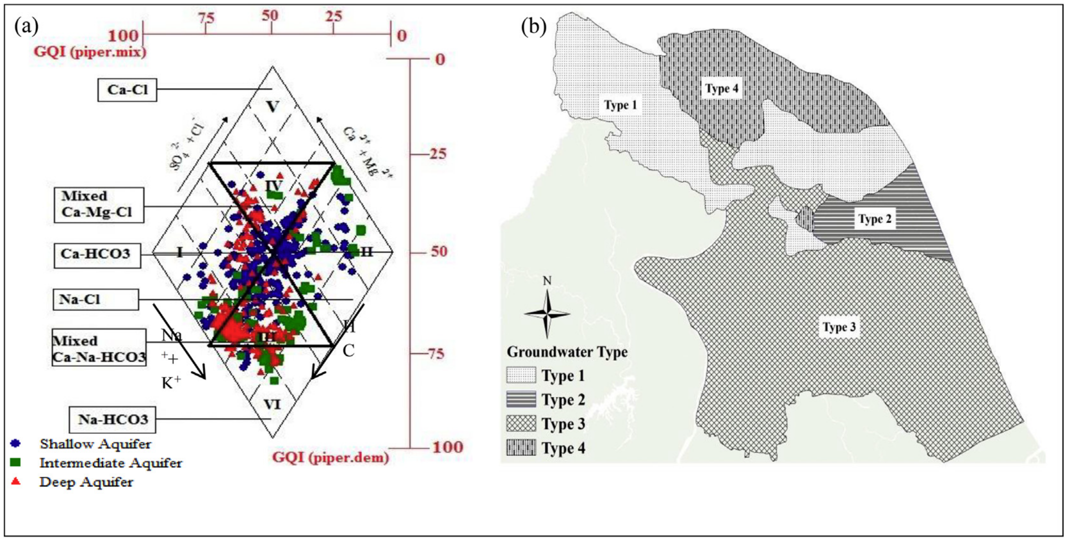

Fig. 4. Representation of groundwater samples from 1989 to 2011 in shallow, intermediate and deep aquifers based on piper domains indexes (a) spatial distribution of groundwater type (b).

of the groundwater was of the Ca-HCO_3 and mixed Ca-Na-HCO_3 types, respectively). Na-Cl (type II) was a dominant groundwater type in the intermediate aquifer, which covered 50% of the groundwater samples. Around 41% of the other samples in the intermediate aquifer comprised of groundwater type III. Groundwater salinity, especially in the coastal areas, can be related to sea water intrusion (Samsudin et al., 2008), and the dissolution of soluble salts in the unsaturated zone. Meanwhile, in the deep aquifer, around 43.7% of the samples encompassed of groundwater type III, while 37.5% of them consisted of type I. The remaining 12.5% comprised of type IV (Ca-Mg-Cl).

Based on the spatial distribution map the majority of the study area (approximately 55.8%) displayed groundwater type III (mixed Ca-Na-HCO3), which covers most of the area in the southern and central Northern Kelantan Basin (Fig. 4b). A limited area of around 54 km^2 (6.1%) of the total area was affected by Na-Cl water type. This could be related to seawater intrusion and trapped fossil seawater in this area which were previously reported by Haryono et al. (1995) and Samsudin et al. (Samsudin et al., 2008). The rest of the areas in north, east-north and eastern parts were covered by groundwater type I (Ca-HCO_3) and type IV (Ca-Mg-Cl).

3.2. Detection of groundwater quality trends

The development of long-term groundwater monitoring data provides a unique opportunity to investigate the processes and factors, which affect groundwater quality in the monitoring area. The trend analysis is a useful method to quantify the long-term variations on the water quality parameters, as well as determining the controlling factors of water quality variations over a specific period of time. In the present study, the Mann-Kendall test was applied on 11 groundwater parameters including, EC, Na, Cl, Ca, Mg, K, HCO_3, NO_3, SO_4, Fe, and Mn. These parameters had been collected 22 years (1989–2011) in the Northern Kelantan Basin (Fig. 5).

It is clearly shown the patterns of the long-term electrical conductivity trends in the sampling wells with respect to the shallow, intermediate, and deep aquifers that the intermediate aquifer had high EC values compared to the EC values of the other aquifers over 22 years. The trend analysis showed that the EC in the shallow and deep aquifer had not been indicating any significant trend (*P*-value, $0.068 > 0.05$ and $0.103 > 0.05$) from 1989 to 2011 (Fig. 5a). In contrast, although the intermediate aquifer showed some fluctuation in its trend over the years, had a decreasing trend in general (*P*-value, $0.044 < 0.05$). However, it was clear from the trend analysis that there had been a sudden increase in the EC values from 1994 to 1999 in the intermediate aquifer. A quick view on the overall time series trends for other groundwater quality parameters revealed the sample patterns of trend variations for Na, Cl, Ca, Mg, and K (Fig. 5a–g). Despite the fact that only Cl, Ca, and Mg showed a decreasing trend in intermediate aquifer, and Na, HCO_3, and K did not show any significant trend from 1989 to 2011, there was a presence of higher concentrations shown by all these anions in intermediate aquifer. The groundwater abstraction was 36.37 million L per day (MLD) in 1981, which increased to 57 MLD in 1990, and led to a rapid increase to 184.35 MLD in 1993 with the installation of 72 new wells in the study area. The results of groundwater over abstraction are clear in Fig. 2, where the groundwater level had decreased sharply from 1995 until 2003 in the intermediate aquifer. Through an over abstraction performed on the fresh groundwater from the intermediate aquifer, the trapped seawater could be pumped and raised up in the aquifer. Subsequently, the concentration of ions in the groundwater samples had been increasing from during 1993–1999 in the Northern Kelantan Basin.

Although, no trend was shown by the long-term variation in iron (Fe) in any of the aquifer layers, an increased concentration could be observed from the iron in both the intermediate and deep aquifers (Fig. 5h). The existence of Fe-bearing minerals including pyrite, siderite, magnetite, and iron silicate could be related to the high concentration of Fe in the deep aquifer in the study area. Similar to the Fe ion, manganese had not been showing any trend in the shallow and intermediate aquifers from 1989 to 2011. However, positive trend could be seen from the manganese in the deep aquifer, which indicated the dissolution of Mn minerals in the sedimentary deposit in the Northern Kelantan Basin (Fig. 5i). In comparison to the trends of other groundwater parameters in the Northern Kelantan Basin, sulfate and nitrate showed different trends, which indicated high concentrations of SO_4 and NO_3 in the shallow aquifer.

The long-term patterns of SO_4 and NO_3 in the groundwater samples had clearly indicated an increasing trend in the shallow aquifer from 1989 to 2011. Furthermore, the annual percentage growth rate for sulfate was around 8.04% in the shallow aquifer. However, no significant trend was shown by the sulfate in the deep and intermediate aquifers (Fig. 5j). There was a possible involvement of multiple chemical process factors as the SO_4 sources in the shallow aquifer. This included sulfate-bearing fertilisers, bacterial oxidation of the sulfur compounds (Papatheodorou et al., 2006), combined with seawater and autotrophic denitrification (Pu et al., 2014).

A significant increasing trend of nitrate concentration in the shallow and intermediate aquifers (Fig. 5k was implied from the pattern of long-

term nitrate variation. In comparison to the intermediate and deep aquifers, the nitrate content in shallow aquifers had a high concentration, which was mostly related to the diffused pollution due to the excessive use an overuse of nitrogen fertilisers on farmlands (Junior et al., 2015). Overall, every year there had been an increase of the nitrate concentration in the groundwater by 5.6% and an increase of it by 7.6% in the shallow and intermediate aquifers from 1989 to 2011.

3.3. Factors controlling the long-term variation of hydrochemistry

The primary factorial explanatory model (model 1) elaborates on the projection of the all variable classes and the samples. In the first two factorial axes, 78.5% of the total data variance is explained. On the other hand, the first and second factors describe 68.7% and 9.68% of total variance, respectively. The first model can be represented by in the correlation between land use activities, geology, years of groundwater sampling, and groundwater hydrochemical variables in the shallow and intermediate aquifers (Table 3). The significant correlations between the marine deposits, the intermediate aquifer, grass and forest lands (no human activities) in the years between 1995 and 2000 are indicated by the highest positive loading factors. These correlations include the ions within the highest class of hydraulic conductivity namely Ca, Na, Cl, Fe, and K, which can be interpreted as the water salinity-hardness factors in association with seawater intrusion. However, the strong negative correlation between the intermediate aquifer and high concentration of sulfate (SO_4^{3}) indicates that sulfate concentrations are relatively low compared to the sulfate concentration of the recent seawater (Samsudin et al., 2008). On the other hand the highest negative loadings belong to the shallow aquifer, palm oil trees (human activity), and acidic intrusion (geology) which are directly correlated to the lowest concentration classes of hydraulic conductivity, Ca, Na, Cl, Fe, and K. they are also associated with the high concentration classes of sulfate. Inverse correlations exist between variables in the first model, and they can indicate two different trends in the groundwater which are controlled by the natural processes and human activities. In the case of the first MCA model, there is no significant factor loading attributed to the groundwater level variations (GWL), rainfall, and nitrate classes. Also, sulfate shows a different factor loading behaviour

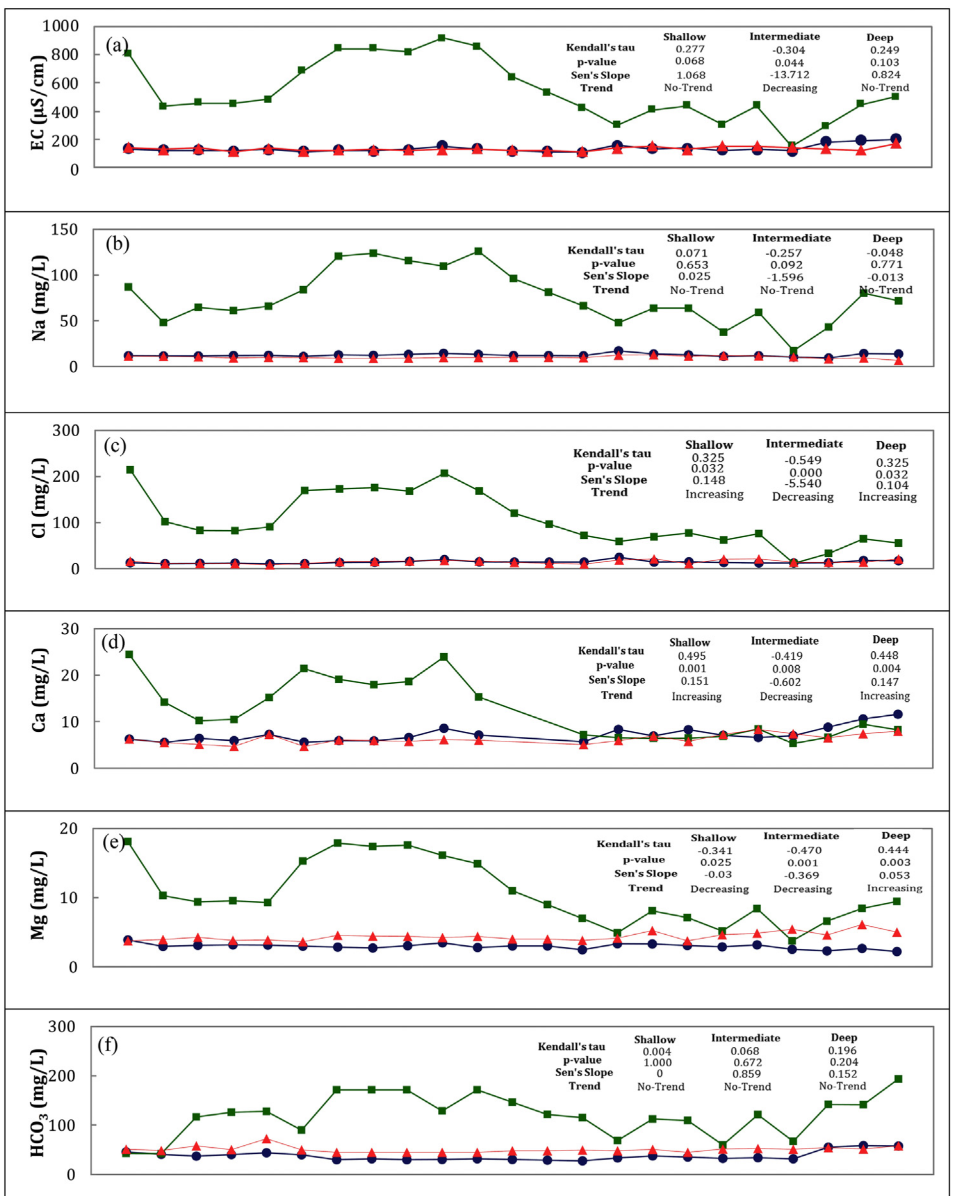

Fig. 5. Groundwater chemistry variations from 1989 to 2011 in the study area.

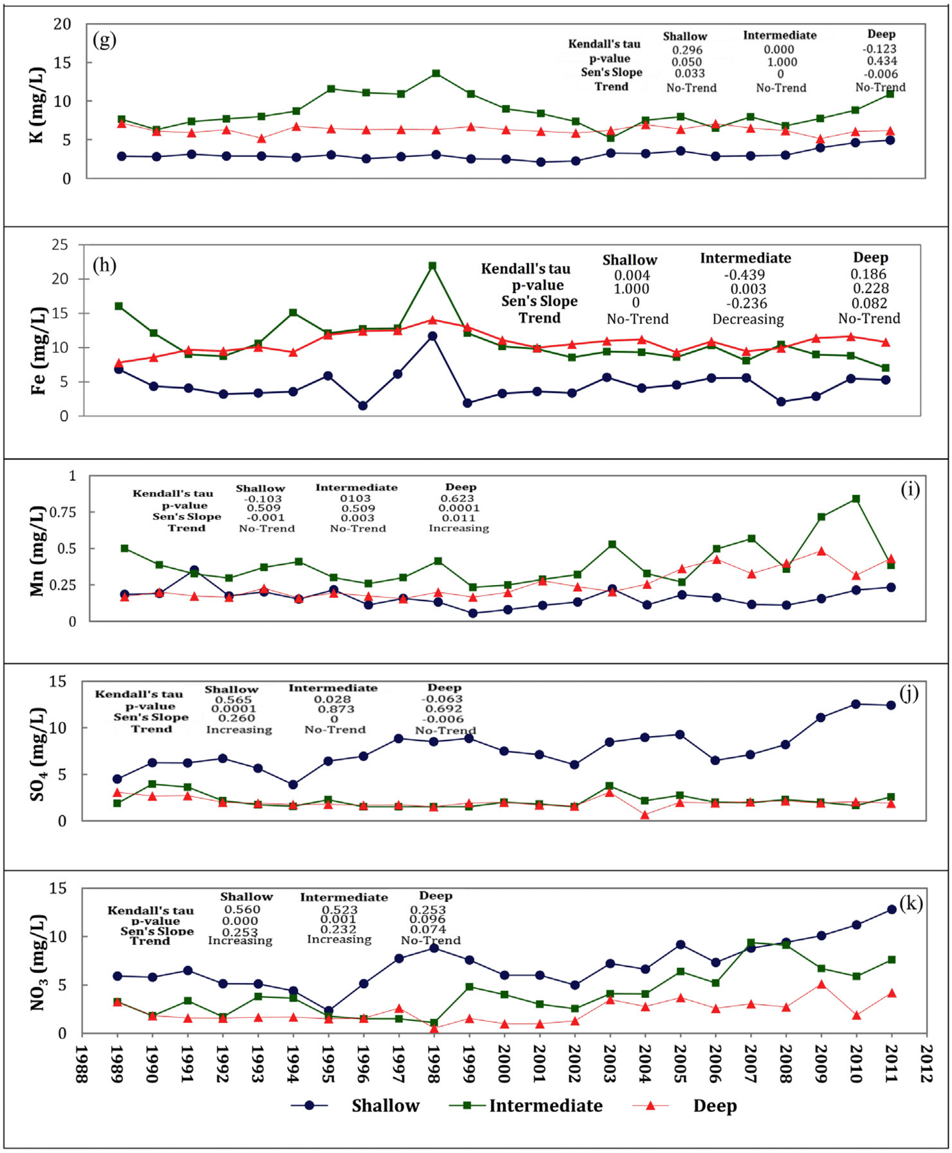

Fig. 5 (*continued*).

compared to the factor loading behaviour of other hydrochemical variables. Therefore, in the second model, NO_3 and SO_4 are skipped from the model, while GWL and rainfall serve as supplementary variables (Table 3). The results indicate an increasing variance of the first two factorial axes, which is by 84%. The active and supplementary variables of model 2 are projected on the primary plane in Fig. 6a. In addition, the highest classes of six active variables (electrical conductivity, Ca, Na, Cl, Fe, and K) are located in the positive side, and correlated with the intermediate aquifer, marine deposit, along with grass and forest land use. Apart from that, the second model clearly presents the role of the geological structure in the chemistry of groundwater in the intermediate aquifer. The existence of ancient seawater which remains trapped during the deposition of sediments within medium to coarse sand can be the main source of hard-brackish water in the intermediate aquifers (Suratman, 1997; 2010). The spatial distribution map of the first factor score (model 2) shows the highest factor loading, which extends along the coastal line (Fig. 6b). The highest factor scores (0.9–1.4) are located in the area that matches to the Na-Cl water type, as shown in Fig. 4b.

The third model was made to build explanatory models for NO_3 based on exclusively non-physicochemical variables, where SO_4, GWL, and rainfall were projected as supplementary variables. The first two axes elaborate on the percentage of the total variance which is 72.1%. This amount shows that the high concentration of NO_3 has a strong correlation with the shallow aquifer, mixed agriculture, palm oil plantations (human activities) from 2001 to 2011 and areas with acidic intrusion (Table 3). The presence of nitrate in groundwater is possibly due to because of the degradation of naturally occurring organic matter or manmade sources such as the nitrogen-fertiliser application, livestock operations, sewage infiltration, and industrial process (Shamsuddin et al., 2016). According to Islami et al. (2012) the application of chemical and natural fertilisers is the factor of the high concentration of nitrate in palm oil plantations. Based on the literatures, a high nitrate concentration is more common in shallow wells, and it generally decreases with the increasing depth of the groundwater (Hu et al., 2005: Rutkoviene et al., 2005).

This is because the nitrogen compounds in the fertilisers are oxidised in aerated soils to soluble nitrate, and the surface-water infiltration can leach from below the root zone into the groundwater (Mahvi et al., 2005). Based on the observation on the projection of a low NO_3 concentration (NO_3^1) with deep aquifer, grass and forest lands (Fig. 6c), it is clearly proven that the high concentration of nitrate attributes to human activities and generally occurs in the shallow aquifer. In addition, a slight correlation to the highest NO_3 class is shown by the class

Table 3
Factor loading of variable classes based on different models.

	Model 1	Model 2	Model 3
Aqui.Sh	**−0.709**	−0.619	**−0.750**
Aqui.Int	**0.877**	**0.895**	0.233
Aqui.De	0.033	−0.073	0.651
LP	0.010	0.011[a]	−0.025[a]
LN	−0.025	−0.029[a]	0.065[a]
RL	−0.131	−0.124[a]	−0.074[a]
RH	0.172	0.155[a]	0.099[a]
Y_1	−0.177	−0.213	0.326
Y_2	**0.652**	0.529	**1.347**
Y_3	−0.194	−0.200	**−0.633**
Y_4	0.152	0.263	**−0.598**
G	**0.966**	**0.858**	**1.567**
C	0.011	−0.013	−0.088
MA	−0.030	−0.062	**−0.632**
Pd	−0.578	−0.288	−0.171
PO	**−0.940**	**−0.976**	**−0.939**
M	**1.204**	**1.185**	**0.885**
CSG	−0.322	−0.357	−0.243
A	**−1.059**	**−1.271**	**−1.394**
NO_3^1	0.227		0.671
NO_3^2	−0.113		−0.218
NO_3^3	−0.237		**−0.822**
SO_4^1	0.467		0.356[a]
SO_4^2	−0.232		0.061[a]
SO_4^3	**−0.679**		−0.473[a]
$COND_1$	**−0.759**	**−0.842**	
$COND_2$	−0.518	−0.522	
$COND_3$	**0.947**	**0.950**	
Ca_1	**−0.708**	**−0.786**	
Ca_2	−0.225	−0.205	
Ca_3	**1.228**	**1.212**	
Na_1	**−0.643**	**−0.724**	
Na_2	−0.529	−0.503	
Na_3	**1.221**	**1.205**	
Cl_1	**−0.687**	**−0.780**	
Cl_2	−0.455	−0.426	
Cl_3	**1.335**	**1.299**	
Fe_1	**−0.637**	**−0.608**	
Fe_2	−0.273	−0.367	
Fe_3	**1.239**	**1.209**	
K_1	**−0.860**	**−0.862**	
K_2	−0.173	−0.240	
K_3	**1.298**	**1.266**	

Significant loadings in bold.
[a] Supplementary factor.

of paddy cultivation (land use activities). The factor of this possibility related to the application of reduced fertilisation for paddy fields, in comparison to the application of reduced fertilisation for oil palm and other cultivations (Islami et al., 2012). Besides, paddy soils, especially in the saturation zone, possess self-purification capabilities for nitrate contamination (Zhu et al., 2003). The spatial distribution map for factor score one in model 3 shows a highly NO_3 contaminated area located in the central and southern regions of the study area, which matches the extension of the agricultural activities within the study area (Fig. 6d). As expected, the lower concentration of nitrate was observed along the coastal line, where natural processes were mostly influenced by it. The weak relation between NO_3^- and the paddy field, despite the large extension of rice cultivation in the study area, could be related to the reduced application of N-fertilisers in the paddy fields and the existence of a self-purification mechanism within the paddy soils in the study area.

3.4. The origin of natural processes

The origin of solutes, hydrochemical processes and mechanisms which generate the groundwater composition can be determined using the compositional relations among dissolved species (Jalali, 2009; Sivasubramanian et al., 2013). The relationship between Na^+-Cl^- has often been used identifying the mechanism for the acquirement of salinity and saline intrusion in coastal regions. A plot of the relationship between Na and Cl (Fig. 7a) indicates a good correlation between Na^+ and Cl in majority of the samples (Fig. 7a). This implies that the groundwater is probably controlled by the water-rock interaction, which is most likely derived from the weathering of calcium magnesium silicate, plagioclase, feldspar and gypsum (Sivasubramanian et al., 2013). Approximately 20% of the samples, mostly comprising of intermediate and deep aquifers, shows ratio values below the seawater ration (0.86) this demonstrates the contamination of groundwater with saline water (Lee and Song, 2007). The values which are close to the seawater ratio line show the recent simple combination of groundwater with seawater (Mercado, 1985). However, around 59% of the samples (mostly from shallow aquifer) were positioned above the theoretical fresh-seawater mixing line, which suggests that the excess of Na^+ is likely due to derived from the silicate weathering of feldspar or plagioclase within the study area (Hussin et al., 2016). Limited samples, which mostly originated from intermediate and deep aquifers show that the low ration values of Na/Cl are especially controlled by the cation exchange process between Na^+ and Ca^{2+}-Mg^{2+}.

The plot of $HCO_3^- + SO_4^{2-}$ and $Ca^{2+} + Mg^{2+}$ (Fig. 7b) shows that majority of the shallow well samples fall under the aquiline. This indicates that the calcite, dolomite, gypsum, and also ion exchange are the dominant reactions in the shallow aquifer system. However, the samples belong to the intermediate and deep aquifers plot above the 1:1 line, which can be the result of a reverse ion exchange. Furthermore, wells numbers 3 and 4 originated from the deep aquifer, while the wells number 12, 13, 14, 31, 32, and 33, were located near the coastal line (Fig. 6c) from the intermediate aquifer with high ratios. Provided if the data point has the tendency towards the right due to excessive $SO_4^{2-} + HCO_3^-$, the ion exchange is the dominant process.

If the ion exchange is the main controlling process of the groundwater composition, the relation between Na-Cl against ($Ca^{2+} + Mg^{2+}$) - ($HCO_3^- + SO_4^{2-}$) will show a negative linear trend with a slope of unity. Fig. 7c, shows a plot with a slope of −0.50, −0.74, and −0.87 for shallow, intermediate and deep aquifers respectively, which implies a high level of ion exchange reaction mostly taking place in the intermediate and deep aquifers.

Some samples spread above and below the linear trend, which suggest that the controlling of groundwater chemistry is not only influenced by the involvement of the ion exchange reaction, it is also influenced by the involvement of other process. Furthermore, the plot of $Ca^{2+} + Ma^{2+}$ versus HCO_3^- shows that majority of samples from the shallow aquifer were positioned around the 1:1 aquiline, which suggests that calcite and dolomite weathering contribute to the transfer of ions to the groundwater (Fig. 7d). Therefore, the major source of Ca^{2+} and Mg^{2+} is the dissolution of carbonate minerals, whereas HCO_3^- originates from silicate weathering and carbonate minerals (Sivasubramanian et al., 2013). Last but not least, the higher ratios (mostly observed in the intermediate and deep aquifers) indicate the extra sources of Ca^{2+} and Mg^{2+} ions balanced by Cl^- and SO_4^{2-} supplied by silicate weathering (Zhang et al., 1995).

4. Conclusions

The current research has revealed that the groundwater chemistry undergoes various transformations induced by natural and anthropogenic hydrological and geochemical processes throughout its evolutionary path in the study area. However, determining the relationship between groundwater quality parameters and types of effective activities and intrinsic factors such as aquifer lithology and groundwater level during long-term monitoring is very complex processes.

The hydrogeochemical investigation in the study area had shown the same hydrochemical facies, namely the Na-HCO_3 and Ca-HCO_3 types in years 1989 and 2011. On the other hand, significant changes in hydrochemical facies were detected in different layers of the aquifer,

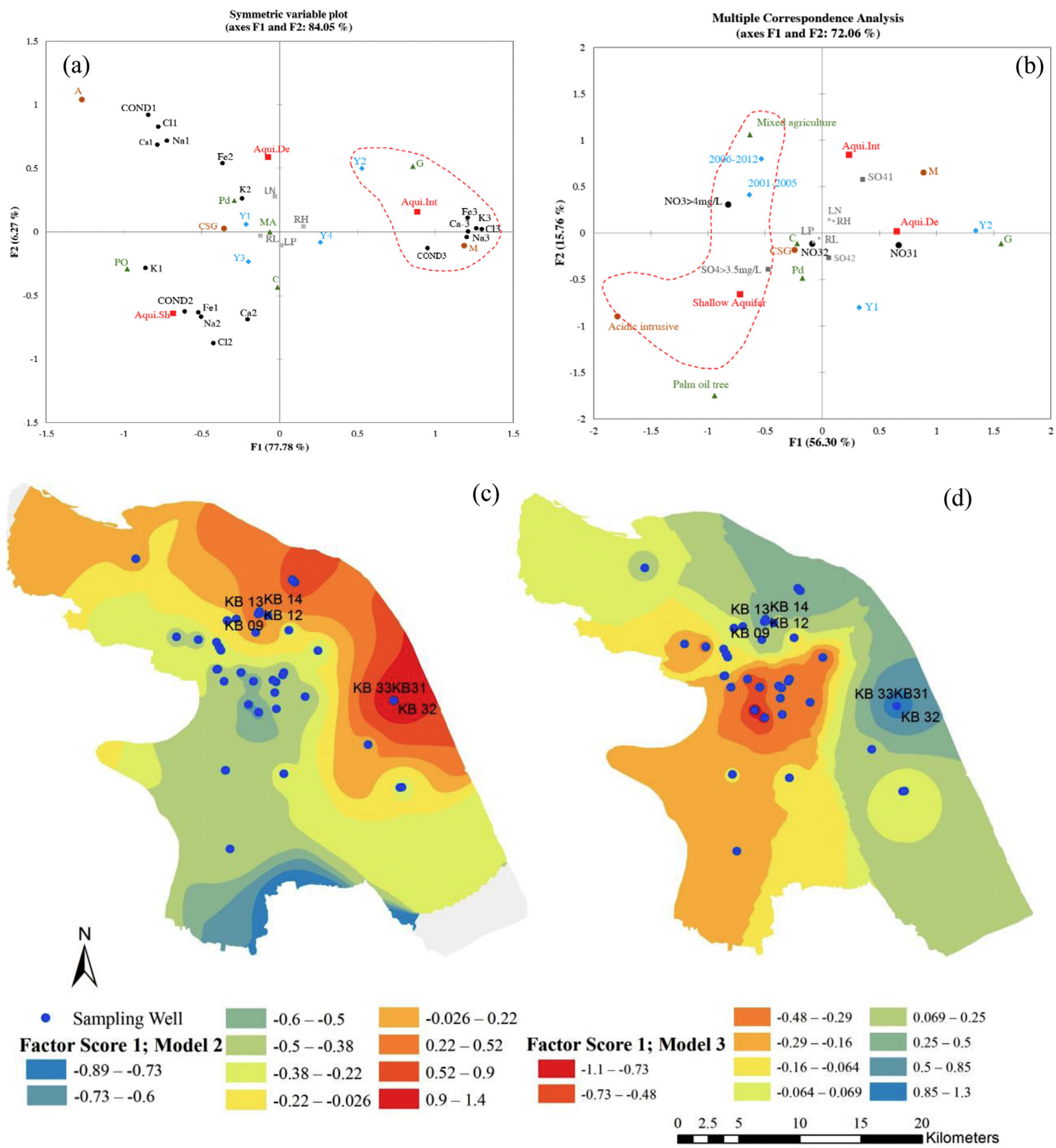

Fig. 6. Primary factorial plane of multiple correspondence analysis (MCA), a) based model 2 include all chemical variables except NO_3, b) model 3 include NO_3 as chemical variable and Spatial distribution of factor score one based on, c) model 2 represents factors influence intermediate aquifer, d) model 3 represents factors influence shallow aquifer.

which varied from Ca-HCO_3 in the shallow to Na-Cl and Ca-Na-HCO_3 in the intermediate and deep aquifers respectively. Similarly, the time series pattern revealed groundwater salinity and high concentration of calcium, magnesium, potassium, iron, and bicarbonate in the intermediate aquifer. On the contrary, the Mann-Kendall test did not show any significant trend for groundwater salinity in the intermediate aquifer. This could imply the role of natural processes as the controlling factors of the groundwater salinity in this layer. In addition, it has been revealed by the MCA revealed that salinisation in the intermediate aquifer is associated with aquifer lithology, specifically by marine deposits. The spatial distribution map constructed by geospatial modeling shows the salinity extension throughout the coastal area, where the existence of ancient seawater trapped between medium to coarse sand sediments was reported by several researchers.

Despite the absence of the prominent trend for the groundwater salinity in the aquifer system, a significantly increasing trend can be seen from the nitrate and sulfate in the shallow aquifer. Given that shallow aquifers are prone to contamination from sub-surface activities, it has been suggested that high concentrations of nitrate and sulfate are resulted from the intensive use of nitrogen-containing fertilisers and contamination with human or animal organic waste. The MCA enhances the understanding that high concentration of nitrate in the shallow aquifer is controlled by land use activities. These are referred to the ones conducted in the area with palm trees and mixed agricultural cultivations, which are mostly expanded in the central and southern sides of the northern Kelantan Basin. Besides, there has been an increasing trend of these cultivations the recent decades. The significant negative correlation between high contamination of nitrate in the shallow aquifer and areas covered with grass and forest indicates the significant effect anthropogenic factors on the shallow aquifer in the study area.

The results of this study provide a scientific view for the decision maker to consider the effects of land use and activities on the factors

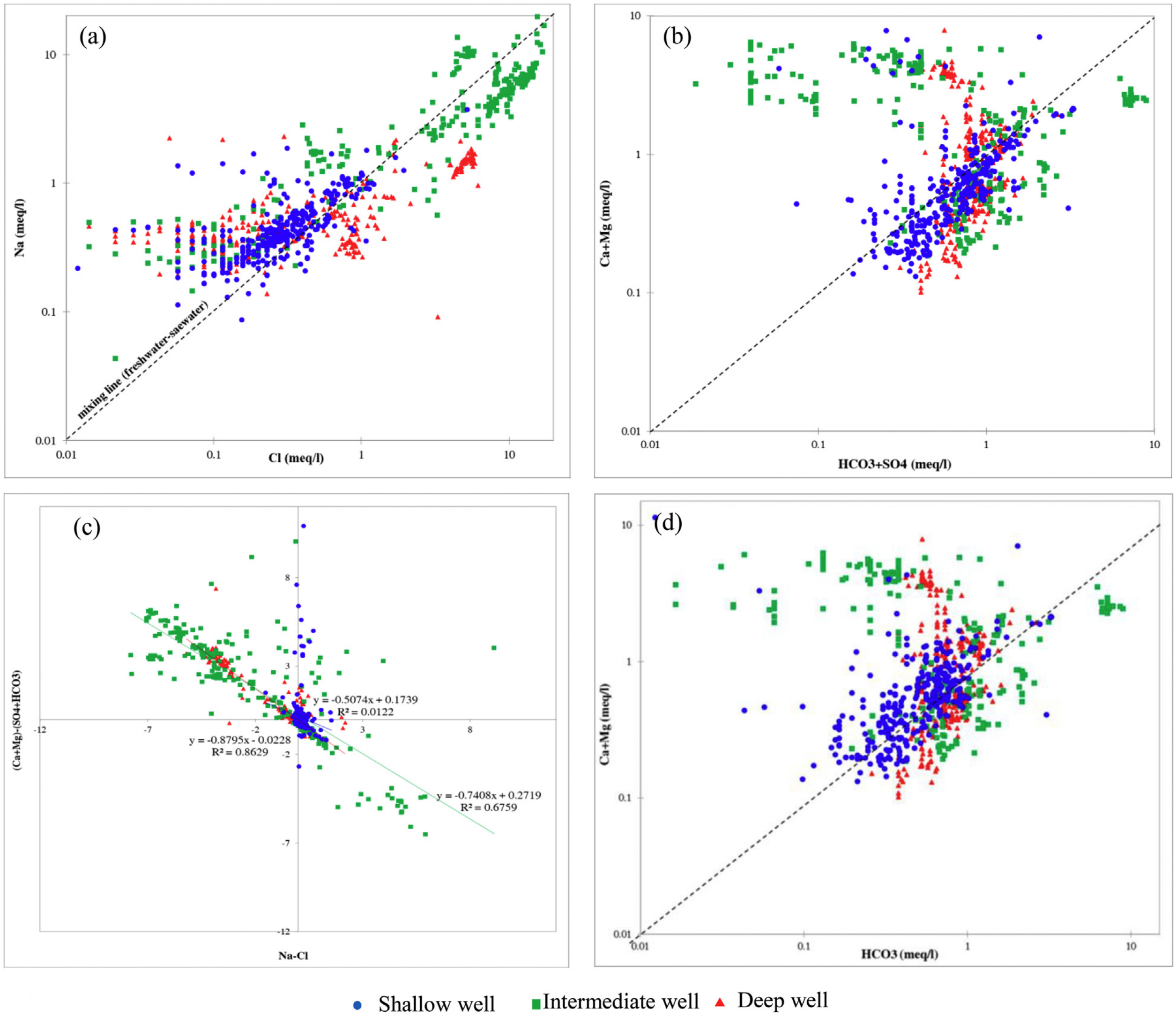

Fig. 7. Distribution of ionic ratio for major ions in shallow, intermediate and deep aquifers.

controlling groundwater quality. Groundwater chemistry has no meaningful correlation with the climate and groundwater level variation data. Nevertheless, future researches which focus on more factors related to water quality such as groundwater extraction values, density of population and run off data would be able to enhance the understanding of the impacts of human activities on groundwater quality depletion.

Acknowledgment

The authors would like to thank the Minerals and Geoscience Department of Malaysia (JMG) and National Hydraulic Research Institute of Malaysia (NAHRIM) for their support in this study. Also, they would like to express their gratitude to the staffs of Faculty of Environmental Studies, Universiti Putra Malaysia for their kind assistance.

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